

Studies on Nanostructure Aluminium Thin Film Coatings Deposited using DC magnetron Sputtering Process

Muralidhar Singh M¹, Vijaya G², Krupashankara M S², B K Sridhara¹, T N Shridhar¹²

¹Department of Mechanical Engineering, National Institute of Engineering, VTU, Mysuru, India

²Department of Mechanical Engineering, R.V.College of Engineering, VTU, Bengaluru, India

Abstract: Nanostructured thin film metallic coatings has become an area of intense research particularly in applications related solar, sensor technologies and many other optical applications such as laser windows, mirrors and reflectors. Thin film metallic coatings were deposited using DC magnetron sputtering process. The deposition rate was varied to study its influence on optical behavior of Aluminum thin films at a different argon flow rate. Studies on the optical response of these nanostructure thin film coatings were characterized using UV-VIS-NIR spectrophotometer with integrating sphere in the wavelength range of (250-2500nm) and Surface morphology were carried out using atomic force microscope with roughness ranging from 2 to 20nm and thickness was measured using Dektak measuring instrument. The reflection behavior of aluminium coatings on polycarbonate substrates has been evaluated. UV-VIS-NIR Spectrophotometer analysis indicates higher reflectance of 96% for all the films in the wavelength range of 250 nm to 2500 nm. Nano indentation study revealed that there was a considerable change in hardness values of the films prepared at different conditions.

Keywords: Thin films, Sputtering, Reflectance, Aluminium

- 1. Introduction** The need for reflectance of light energy and heat energy is continually increasing due to the global rapid growing population, industrial development, and domestic needs. The reflectance of solar energy can reduce use of nonrenewable energy sources such as fossil fuels, petroleum fuels, and fissionable minerals [1]. Depending upon application thin films are categorized into optical, electrical, magnetic, chemical, mechanical and thermal coatings. Reflectance of solar energy is used in solar thermal devices such as solar cookers, for cooking, water heating, space heating, space cooling, and heat generation process. Solar cookers are easy to build, are smoke free, nonpollutant [2]. Aluminum is the most widely used non-ferrous metal. It is low cost and has a unique set of fine material properties that have made it ideal to be used in many common applications; such as in structural components and mirror reflectors. Aluminum is one of the most abundant metals found on Earth, and the recent advancement in the efficiency of extracting aluminum from ore has made it relatively cheap to procure. It has about one-third the density and stiffness of steel; it is ductile [3].The reflectivity or reflectance (R), of a surface is an



intrinsic optical property of a surface. In many optical, electrooptic, telecommunications, solar concentrator and architectural applications, reflectance must either be controlled aluminum thin films are used to reflect infrared wavelengths to reduce heat loss or ingress through windows. Infrared reflectance must be maximized while keeping visible light transmission through the window high. Concentrating collectors make use of curved reflectors to concentrate sunlight on a receiver. The intensity of sunlight falling on the receiver can be up to 60 times the intensity of normal sunlight [4-5]. The sputtering or sputter erosion of solid material by positive-ion bombardment is widely used as a source of vapor for thin film deposition because of unique combination of advantages over other techniques such as any materials can be volatilized, compounds are volatilized stoichiometrically, and film deposition rate can be made uniform over very large areas. To ensure the good performance and long technical life time of the reflectors the reflective aluminium thin film coatings was analyzed in this work, the optical and mechanical properties of the films was assessed.

It is the purpose of this paper to report on new measurements of the optical reflectance and mechanical properties of sputtered aluminum thin films produced from purest aluminum with extremely high deposition rate on polycarbonate substrate, and to discuss in addition the effect of argon flow and deposition power on the reflectance of aluminum in UV Vis NIR region and mechanical properties of aluminum thin films.

- Material and Methods** Aluminum thin film coatings were deposited on polycarbonate substrates using DC magnetron sputtering process. The substrates were cleaned followed by de ionized water rinse followed by dry nitrogen gas purge figure 1 shows the schematic diagram of the sputtering chamber. DC magnetron sputter coating processes are carried out in vacuum chamber at a base pressure of 1×10^{-5} mba using argon gas (99.999%), were performed at 500,1250 and 2000W by using DC power supply. The deposition chamber was filled with pure argon gas with a flow rate of 200, 350 and 500sccm for target cleaning; pre sputtering process was carried out with shutter positioned over the target there by shielding the flow of plasma towards the substrate for about 5 minutes. The deposition conditions are given in table 1 all depositions were performed at a fixed substrate to source distance of 270mm with a substrate rotation at a rate of 2rpm The substrate temperatures were maintained at room temperature. The coating thickness was maintained constant at 400 nm for all the samples.

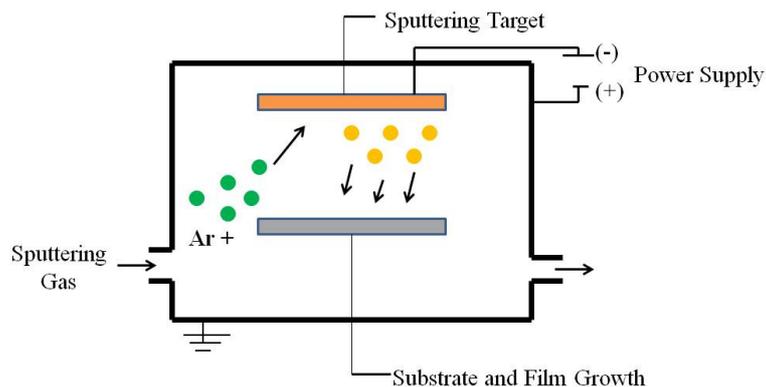


Figure 1 Schematic of the Pulsed DC Magnetron Sputtering Setup

The coatings were characterized for optical reflection measurements on the films were carried out using UV-VIS-NIR spectrophotometer (Model Lambda 750) with the wavelength ranging between 250-2500 nm. Film thickness measurements and surface topography of the films were carried out using DEKTAK 6M surface profiler. Surface topography and mechanical properties of the films of the thin films were done using a nanoindenter (Hysitron-Make).

Table 1 Sputtering parameters for aluminum thin film deposition

Process Parameters	Values
Sputtering Gas	Argon
Argon flow rate (sccm)	200, 350, 500
Power supply	DC
Sputtering Power (w)	500, 1250 and 2000
Temperature	RT
Substrate	Polycarbonate
Coating Thickness (nm)	400
Base Pressure (torr)	1×10^{-5}
Target	Aluminum

Hardness and elastic modulus measurements are the two mechanical properties measured most frequently using indentation techniques are the hardness, H, and the elastic modulus, E. As the indenter is pressed into the sample, both elastic and plastic deformation occurs, which results in the formation of a hardness impression conforming to the shape of the indenter. During indenter withdrawal, only the elastic portion of the displacement is recovered, which facilitates the use of an elastic solution in modeling the contact process [10,11,12]. Figure 2 (a) shows a typical load–displacement curve and the deformation pattern of an elastic–plastic sample during and after indentation. In Figure 2 (b), h_{max} represents the displacement at the peak load, P_{max} . h_c is the contact depth and is defined as the depth of the indenter in contact with the sample under load. h_f is the final displacement after complete unloading. S is the initial unloading contact stiffness. Nanoindentation hardness is defined as the indentation load divided by the projected contact area of the indentation. It is the mean pressure that a material can support under load. From the load–displacement curve, hardness can be obtained at the peak load as

$$H = \frac{P_{max}}{A} \tag{1}$$

where A is the projected contact area. Measurement of the projected contact area from a load–displacement curve requires the contact depth, h_c . The elastic modulus of the indented sample can be inferred from the initial unloading contact stiffness,

$$S = \frac{dP}{dh} \tag{2}$$

i.e. the slope of the initial portion of the unloading curve. Based on relationships developed by Sneddon [13]

E_r is the reduced elastic modulus, which accounts for the fact that elastic deformation occurs in both the sample and the indenter. E_r is given by

$$\frac{1}{E_r} = \frac{1-\nu^2}{E} + \frac{1-\nu_i^2}{E_i} \tag{3}$$

where E and ν are the elastic modulus and Poisson’s ratio for the sample, respectively, and E_i and ν_i are the same quantities for the indenter. For diamond, $E_i = 1141$ GPa and $\nu_i = 0.07$ [10, 13].

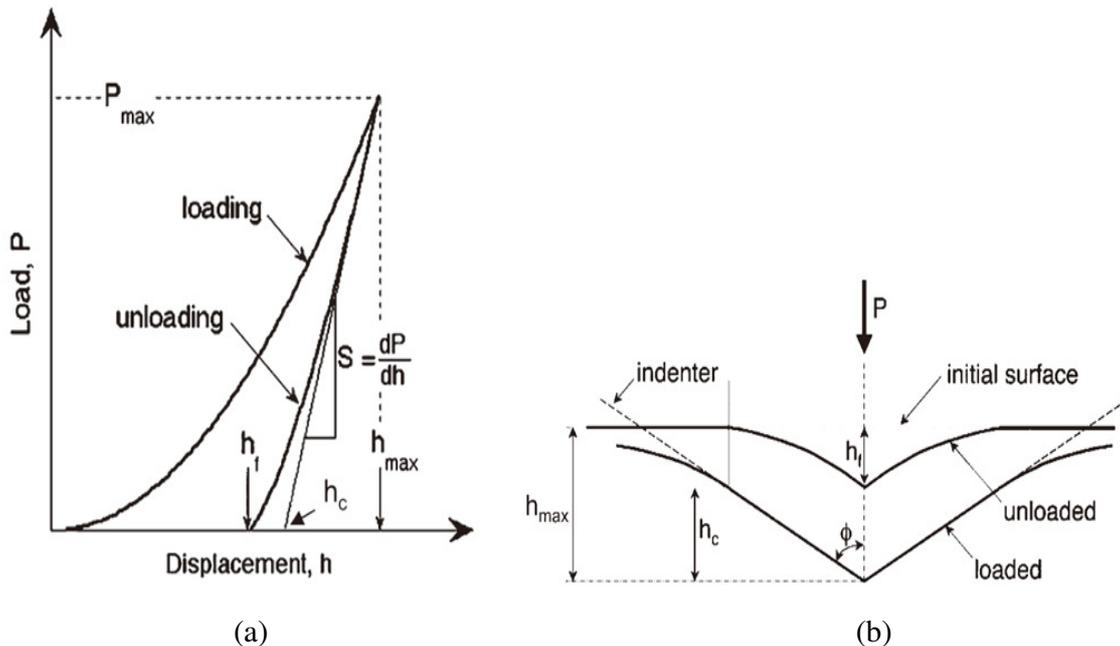


Figure 2 (a) A typical load–displacement curve and (b) the deformation pattern of an elastic – Plastic sample during and after indentation [11]

3. **Results and Discussions** Rate of deposition with respect to argon flow and processing pressure curves of the aluminium thin film coatings are shown in figure 1(a) and current and voltage variations are shown in figure 1(b).

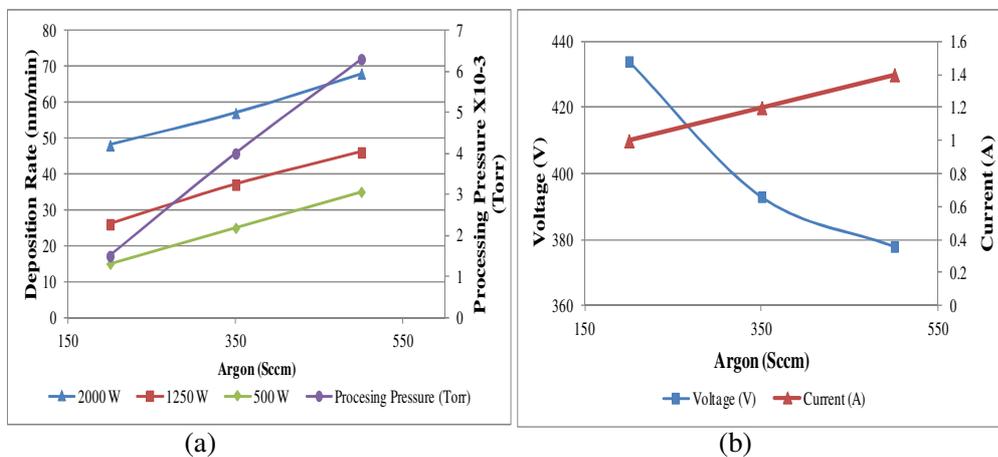


Figure 3 (a) Rate of deposition with respect to Argon Flow and processing Pressure (b) Current Vs Voltage Variation With respect to Argon

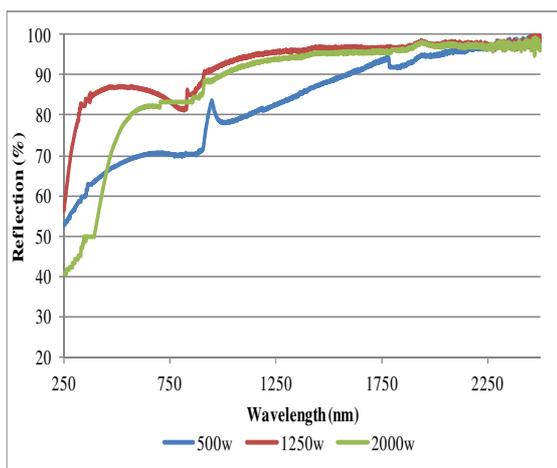
As mentioned earlier, aluminium has a broad spectral band of high reflectivity and therefore, it is not surprising to see that one of its prevailing uses is in solar reflector coatings where a thin film, typically in the order of nanometer, of pure aluminium is deposited on a substrate by DC magnetron sputtering in vacuum. Figure 3 (a) shows the thickness variation 12nm to 13nm at 200sccm argon flow with processing pressure of 1×10^{-3} torr, 23nm to 46nm at 350sccm argon flow with processing pressure of 4×10^{-3} torr and 48nm to 69nm at 500 sccm argon flow with processing pressure of 6.2×10^{-3} torr and process pressure variation is as shown figure 3 (b)

shows the current and voltage variation with respect to change in argon flow. Pure aluminium reflectors are not protected with an overcoat. It is prone to oxidation tarnishing, and a barrier oxide layer is formed when aluminium is reacting with oxygen in air. The resulting oxide layer is tough and corrosion resistant in normal environments. However, it can become susceptible to changing environmental conditions and cannot be easily cleaned without damaging the aluminium coating. In addition, even though oxidation reduces aluminium reflectance by creating unnecessary scattering, the effect is quite limited in the visible spectrum. Therefore, for low end consumer and non-critical applications, the cost benefits of using just bare aluminium can significantly outweigh the marginal reflectance improvement in visible spectrum. The average reflectivity for bare aluminium is above 96% for 400 nanometers at different powers. Because it is also not coated, it is not sensitive to polarization and angle of incidence and so provides consistent high reflectance throughout the near-ultraviolet, visible and near-infrared regions as shown in figure 4.

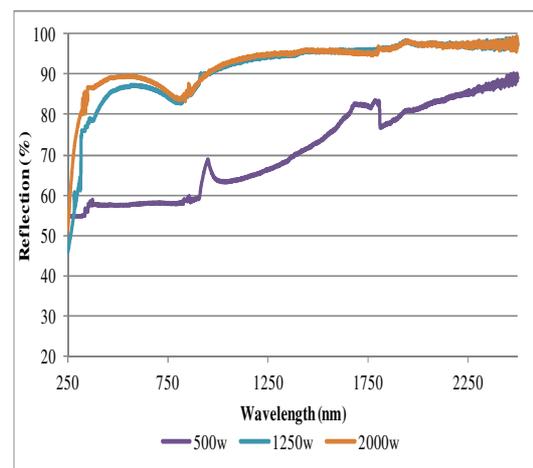
UV-VIS-NIR spectra of the Aluminium thin films at different power and at different argon flow are shown in Figure 4. Table 4 shows the percentage distribution of reflection in UV-VIS-NIR regions. There is a linear increase in reflectance from UV to NIR and a corresponding linear in reflection in the wavelength range of 250 to 2500nm. However, from 1500-2500nm, there is a significant difference between the two plots since the surface roughness, hardness and film stress begin to dominate in the NIR region and hence there is constant reflection. There is no significant change in the response of UV-VIS- NIR of films varied with different power and argon flow. From the reflection spectra there is a clear indication that the Aluminium acts as perfect reflector.

Table 2 Optical reflectance of aluminium films deposited on polycarbonate substrate

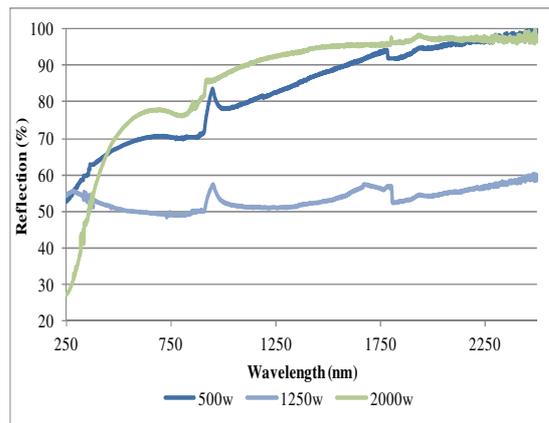
Argon flow(sccm)	200			350			500		
	500	1250	2000	500	1250	2000	500	1250	2000
UV (100-380)	57	75	45	56	66	77	55	60	40
VIS (380-780)	59	86	76	60	86	88	51	69	73
NIR (780-2500)	90	96	96	78	96	96	55	92	96



(a)



(b)

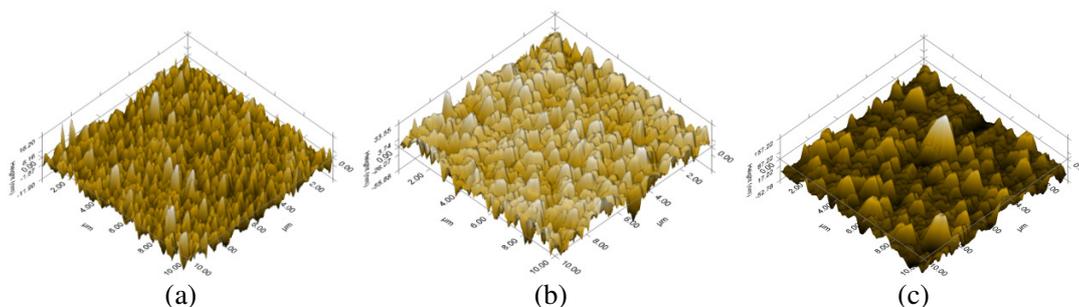


(c)

Figure 4 Shows Reflectance spectra for argon flow at (a) 200sccm (b) 350sccm and (c) 500sccm

Optical Reflectance of Aluminium Films the percentage of optical reflectance of aluminium films deposited on polycarbonate substrates at various wave lengths is shown in Table 2. The optical requirement that must be fulfilled for reflectors is high reflectance in the entire wavelength range of the solar spectrum. Metal, that obey the high reflectance is aluminium, should therefore be suitable. Aluminium is the best candidate, with a solar hemispherical reflectance of 92% for aluminium. figure 4(a) shows the reflection spectra obtained from DC magnetron sputtering process at room temperature. High reflection property is obtained when the argon flow 350 sccm at 2000w the reflection in UV region 77% , VIS region 88% and NIR region 96%, we have achieve optimum thickness of 400 nm. If the power is increases too much, the aluminium layer will reflection remains same due to the higher dissolving power. Moreover, with increase in argon flow 200 sccm to 500 sccm the reflection increases from 50% in UV region to 55 % and the optical properties of the aluminium films will 96% with change in argon flow and power. The optimum reflection can be achieved by maintaining argon flow of 500 sccm with sputtering power of 2000w i.e., 96% reflection.

Figure 5 shows the AFM images of the samples with varying argon flow and power surface roughness of the aluminium thin film coatings at 200sccm argon flow the roughness increased from 3nm to 17nm with increase in power from 500 to 2000w shown in figure 5 (a), (b) and (c), at 350sccm argon flow the roughness increased from 8nm to 13nm with increase in power from 500 to 200w figure 5 (d), (e) and (f), and at 500sccm argon flow the roughness increased from 4nm to 16nm with increase in power from 500 to 2000w as shown figure 5 (g), (h) and (i) and table 3 shows the variations in roughness with increase in argon flow and power.



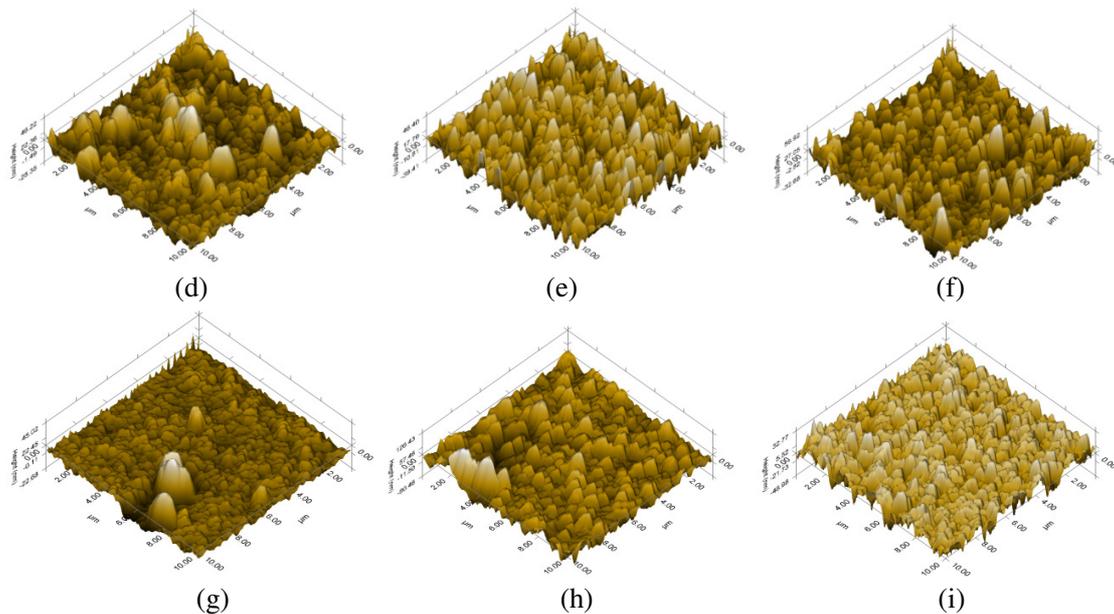


Figure 5 AFM images of Aluminium thin films (a) at 200 Sccm & 500W (b) 200 Sccm & 1250W (c) 200 Sccm & 2000W (d) at 350 Sccm & 500W (e) 350 Sccm & 1250W (f) 350 Sccm & 2000W (g) 500 Sccm & 500W (h) at 500 Sccm & 1250W (i) 500 Sccm & 2000W

Table 3 Optical reflectance of aluminium films deposited on polycarbonate substrate

Argon flow(sccm)	200			350			500		
Power (W)	500	1250	2000	500	1250	2000	500	1250	2000
Ra(nm)	2.8	9.6	16.6	7.1	10.2	12.3	3.9	13.2	16.3
Rq(RMS)	5.5	12.4	19.7	9.5	13.8	15.4	6.4	17.1	19.5

Figure 6 shows the nano indentation plots of DC magnetron sputter coated aluminium thin films with argon flow at (a) 200sccm (b) 350sccm and (c) 500sccm . The hardness of the aluminium films coated at 200sccm as decreases from 1.7 to 0.4Gpa as the power increases from 500 to 2000w, at 350sccm the hardness increases from 1.8 to 0.5Gpa while that of coatings deposited with at 500sccm has a hardness of 1.9GPa due to the effect of surface roughness. The stiffness and modulus values of these films are given in Table 3. There is a slight variation in the stiffness while the modulus is varied between 10 to 20%.

Table 4 Nano indentation mechanical property of aluminium thin film at (a) 200 Sccm (b) 350 Sccm and (c) 500 Sccm

Argon flow(sccm)	200			350			500		
Power (w)	500	1250	2000	500	1250	2000	500	1250	2000
Stiffness (µN/nm)	5.6	6.6	7.1	5.4	6.3	6.5	5.5	7.4	8.1
Hardness(GPa)	1.7	0.5	0.4	1.8	0.6	0.5	1.9	0.7	0.6
Modulus(GPa)	9.7	6.1	5.7	9.4	6.2	5.9	9.2	6.5	6.2

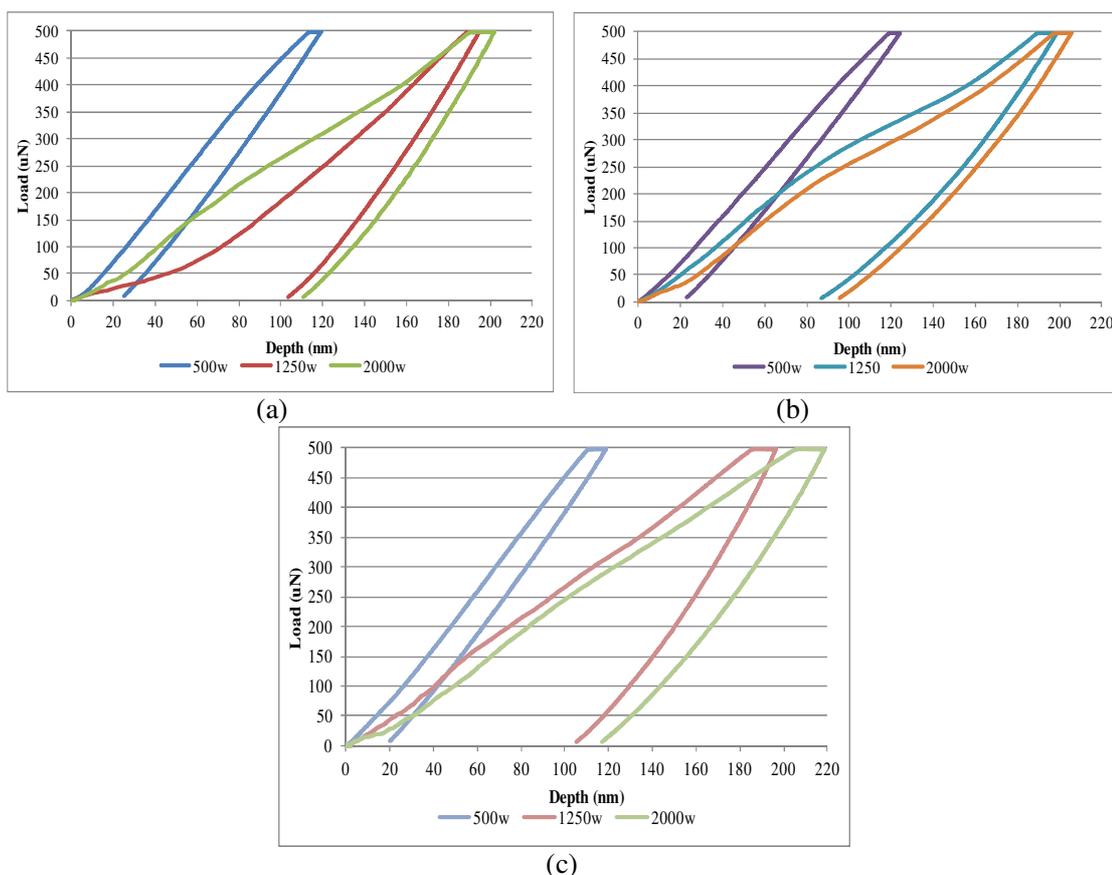


Figure 6 Nanoindentation test Aluminium films at a maximum load of 500uN (a) 200 Sccm (b) 350 Sccm and (c) 500 Sccm

4. Conclusions

DC magnetron sputter deposition of thin films of aluminium on polycarbonate substrates results in formation of pure Aluminium films if the substrate temperature is at RT; if the Argon flow and power is varied from 200 to 500sccm with power ranging between 500 to 2000w, even though the sputter target is 4N pure Aluminium, the as deposited thin film on the substrate shows variation in optical and mechanical properties. The UV-VIS-NIR responses of the coatings deposited at different flow rate and power are similar in the wavelength range of 250 to 2500 nm. With increase in power from 500w to 2000w with argon flow rate of 500sccm, had lower surface roughness ($R_a < 3$ nm) compared to the coating deposited with argon flow rate at 500sccm. When the argon flow rate was maintained at 500sccm during sputter deposition of 4N purity aluminium, as a result the coating had a hardness of 1.9GPa using DC power supply, while that of pure aluminium deposited at 200sccm had a hardness of 1.7GPa. Hardness of the aluminium thin films with variable power is reduced.

5. Acknowledgements

The authors would like to thank the Solar Energy Research Initiative (SERI) Department of Science and Technology, New Delhi, India for funding this research work. The authors gratefully

acknowledge and the support the management of R.V. College of Engineering, Bangalore, The National Institute of Engineering Mysuru, and Visveswaraya Technological University, Belagavi.

6. References

1. J.Ligot, S. Benayoun, J.J. Hantzpergue, J.C. Remy, *Solid-State Electronics*,1075-1078,43 (1999).
2. J. Li, D. R. Alexander, H. Zhang, *Optics Express*, 1939–1945, 15 (2007).
3. Ron Liu, *Optics Express*, 521-27, 17, 2009.
4. Kalogirou S. A, *Application Energy*, 337–61,76(2003).
5. Kalogirou, S. A , *Application energy*, 231–295,77(2004).
6. C. E. Kennedy, R.V. Smilgys, D. S. Kirkpatric, J. S. Ross, *Thin Solid Films*, 303-309, 304, (1997).
7. Thomas Fend, Bernhard Hoffschmidt, Gary Jorgensen, Harald Kuster, Dirk Kruer, Robert Pitz-Paal, Peter Rietbrock, Klaus-Jurgen Riffelmann, *Solar Energy*, 149–155,74 (2003).
8. Maria Brogrena, Anna Helgesson, Bjorn Karlsson, Johan Nilsson, Arne Roos, *Solar Energy Materials & Solar Cells*, 387–412,82,(2004).
9. Maria Brogren, Bjorn Karlsson, Arne Roos, Anna Werner, *Solar Energy Materials & Solar Cells*, 491–515,82(2004).
10. Bhushan B. *Handbook of micro/nanotribology*. 2nd ed. Boca Raton (FL): CRC Press, (1999).
11. Huang, Y M, Y Li, K He, and C X Pan., *Materials Science and Technology*, 1453-1460, 27 (2011).
12. Tabor D, *Philos Mag A*, 1207–21, 74 (1996).
13. Pethica J B, Hutchings R, Oliver WC. , 593–606,48 (1983).
14. Oliver WC, Pharr GM., *Journal Material Research*, 1564– 83, 7(1992).